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Cross section measurements of the reactions induced by deuteron particles on ^{13}C

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Abstract

Nuclear reactions induced by deuterons have been found to be an ideal analysis tool for depth profiling of light elements in the first microns of materials. In particular, the nonresonant nuclear reactions (d, p), (d, α) and (d, t) are well adapted to determine depth distributions of ^{12}C and ^{13}C in a single measurement. Nevertheless, only the cross section of the $^{12}\text{C}(\text{d}, \text{p}_0)^{13}\text{C}$ nuclear reaction is well known for various experimental configurations. Thus, we measured the differential cross sections of the $^{13}\text{C}(\text{d}, \text{p}_0)^{14}\text{C}$, $^{13}\text{C}(\text{d}, \alpha_0)^{11}\text{B}$, $^{13}\text{C}(\text{d}, \alpha_1)^{11}\text{B}$ and $^{13}\text{C}(\text{d}, \text{t}_0)^{12}\text{C}$ nuclear reactions. A thin ^{13}C foil (83 nm thick) was used and the measurements were performed at deuteron energies from 0.5 to 1.65 MeV for different laboratory angles of detection (135° , 150° and 165° with respect to the incident beam). Then, the results obtained in this work were compared to cross sections measured by Marion and Weber for a detection angle of 135° .

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1. Introduction

Ion beam analysis has been applied for many years to measure carbon concentrations and depth profiles, and to perform stable isotopic tracing using materials enriched in ^{13}C . The use of the narrow resonance ($\Gamma = 75$ eV) at 1748 keV of $^{13}\text{C}(\text{p}, \gamma)^{14}\text{N}$ reaction is particularly well adapted to obtain ^{13}C areal densities and profiles with high depth resolution [1]. Nevertheless this method does not allow to determine depth distributions of other carbon isotopes with a single measurement. In that case, the nonresonant nuclear reactions (d, p), (d, α) and (d, t) are required. Indeed, proton, alpha and triton particles are emitted at different energies from $^{12}\text{C}(\text{d}, \text{p}_0)^{13}\text{C}$, $^{13}\text{C}(\text{d}, \text{p}_0)^{14}\text{C}$,

$^{13}\text{C}(\text{d}, \alpha_0)^{11}\text{B}$, $^{13}\text{C}(\text{d}, \alpha_1)^{11}\text{B}$ and $^{13}\text{C}(\text{d}, \text{t}_0)^{12}\text{C}$ reactions. Thus, despite a poorer depth resolution than that available with resonance techniques, these nuclear reactions can be used to depth profile ^{12}C and ^{13}C simultaneously.

The exploitation of these nuclear reactions to depth profile light elements requires the use of accurate cross sections. The $^{12}\text{C}(\text{d}, \text{p}_0)^{13}\text{C}$ cross section is well known at deuteron energies from 0.8 to 1.1 MeV and has been recently measured again [2], which is not the case for nuclear reactions induced by deuterons on ^{13}C [3,4]. Thus, differential cross sections of the $^{13}\text{C}(\text{d}, \text{p}_0)^{14}\text{C}$, $^{13}\text{C}(\text{d}, \alpha_0)^{11}\text{B}$, $^{13}\text{C}(\text{d}, \alpha_1)^{11}\text{B}$ and $^{13}\text{C}(\text{d}, \text{t}_0)^{12}\text{C}$ reactions were measured in this energy range, using a thin self-supported ^{13}C foil. The composition and the thickness of this foil were checked by Rutherford Backscattering Spectroscopy (RBS) and profilometry. The well known carbon build-up phenomenon occurring during beam irradiation [5] was determined and taken into account in this work. Finally, our results were compared to the ones obtained fifty years ago by Marion and Weber [3,4].

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2. Experimental

Three silicon surface barrier detectors (700 μm depleted zone) were used for the detection of charged particles emitted by nuclear reactions. The detectors were respectively positioned at 135° , 150° and 165° with respect to the incident beam (solid angle of ~ 2.1 msr). The RBS and NRA analyses were performed, without any absorber, in this geometry with ALTAIS,¹ a 2 MV Tandetron accelerator installed at the University of Namur (Belgium). The incident beam had a 1 mm diameter section on the target and the vacuum pressure did not exceed 5×10^{-6} Pa. The uncertainty on the accelerator energy was less than 200 eV [6].

The thin ^{13}C foil used for cross section measurements was provided by ACF metals. The thickness given by the manufacturer (82 ± 8 nm) was checked by profilometry using a Surface Profile Measuring System Dektak from Veeco Instruments. Its composition was determined by RBS using a 1 MeV ^4He beam. The incident current was approximately 20 nA. This method may only be used for incident α energies lower than 2 MeV, because above this energy, deviations with respect to Rutherford backscattering become too significant [7]. A very thin film of gold was deposited on the ^{13}C foil by magnetron sputtering technique. The deposition time was controlled in order to obtain a film of about 30×10^{15} at cm^{-2} and its exact thickness ($32.2 \pm 0.6 \times 10^{15}$ at cm^{-2}) was determined by RBS using a 1 MeV ^4He beam. Then, the number of incident particles N_i and the solid angle of detection Ω , were determined using the following equation:

$$N_i \times \Omega = \frac{A_{\text{Au}}}{N_{\text{Au}} \times \left(\frac{d\sigma}{d\Omega}\right)_{\text{Au}}}, \quad (1)$$

where A_{Au} is the number of particles backscattered by gold nuclei (area of the gold peak), N_{Au} is the areal density of gold (at cm^{-2}) and $\left(\frac{d\sigma}{d\Omega}\right)_{\text{Au}}$ is the theoretical elastic cross section for gold obtained by Rutherford formula.

NRA measurements were performed at deuteron energies ranging from 0.5 to 1.65 MeV by 25 or 50 keV energy steps. The incident current was approximately 250 nA. A Faraday cup positioned behind the thin ^{13}C foil allowed us to measure the beam current in order to monitor the time of each acquisition. A ring biased at -150 V was placed between the thin carbon foil and the faraday cup in order to prevent secondary electrons to escape. The charge integrated into the faraday cup was 100 μC for each energy step. By using the $N_i \times \Omega$ product given by Eq. (1), the differential cross sections of $^{13}\text{C}(d, p_0)^{14}\text{C}$, $^{13}\text{C}(d, \alpha_0)^{11}\text{B}$, $^{13}\text{C}(d, \alpha_1)^{11}\text{B}$, $^{13}\text{C}(d, t_0)^{12}\text{C}$ reactions were determined by the following relation:

$$\left(\frac{d\sigma}{d\Omega}\right) = \frac{A_C}{A_{\text{Au}}} \frac{N_{\text{Au}}}{N_C} \left(\frac{d\sigma}{d\Omega}\right)_{\text{Au}}, \quad (2)$$

where A_C is the number of emitted particles by the nuclear reaction under study and N_C is the ^{13}C areal density of the thin foil (at cm^{-2}). This technique of gold marker is very effective to decrease the error made on the measurement of the solid angle of detection and the number of incident particles.

3. Results and discussion

The composition of the thin ^{13}C foil was determined from the simulations of experimental RBS spectra performed with SIMNRA code (Table 1) [8]. These measurements show the presence of contamination layers on both sides of the foil. These contamination layers (thickness of 3×10^{16} at cm^{-2}) are composed of about 92 at% of carbon and 8 at% of oxygen. The ^{12}C and ^{16}O isotopes were also detected in the bulk of the foil, at respectively, 1.5 at% and 0.5 at%. Finally, the ^{13}C areal density of the thin foil was determined to be $(8.63 \pm 0.22) \times 10^{17}$ at cm^{-2} which corresponds to an energy loss of 6 keV for a 1 MeV deuteron beam. The measured thickness (83 ± 2 nm) corresponds to the one determined by profilometry (89 ± 8 nm) and given by the manufacturer (82 ± 8 nm).

A typical spectrum obtained at 165° with 1.05 MeV deuteron particles is shown in Fig. 1. The very intense peak

Table 1

Composition of the thin ^{13}C foil according to simulations of experimental RBS spectra performed with SIMNRA code

Thickness of layers (10^{15} at cm^{-2})	32.2	30	880	30
Composition (at%)				
^{198}Au	100	0	0	0
^{13}C	0	1	98	1
^{12}C	0	91	1.5	91
^{16}O	0	8	0.5	8

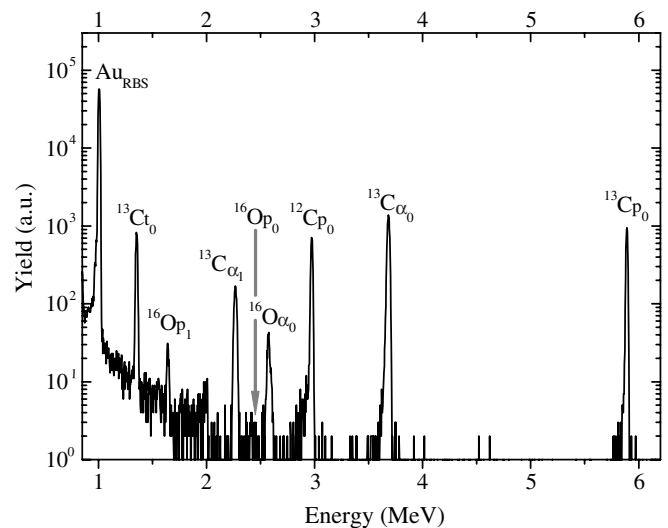


Fig. 1. Typical NRA spectrum obtained at 165° (lab) by bombardment of 1.05 MeV deuteron particles on the thin ^{13}C foil with areal density of $(8.63 \pm 0.22) \times 10^{17}$ at cm^{-2} . The incident current was approximately 250 nA and the acquisition time was about 400 s (integrated charge of 100 μC).

¹ Accélérateur Linéaire Tandetron pour l'Analyse et l'Implantation des Solides.

labeled Au_{RBS} and observed at 1 MeV corresponds to the deuteron particles backscattered by thin film of gold. The peaks labeled $^{13}\text{Cp}_0$ and $^{13}\text{C}\alpha_0$ are due to, respectively, $^{13}\text{C}(\text{d}, \text{p}_0)^{14}\text{C}$ and $^{13}\text{C}(\text{d}, \alpha_0)^{11}\text{B}$ nuclear reactions. The same notation is adopted for all the reactions induced by deuterons on other elements. The $^{13}\text{C}(\text{d}, \text{p}_0)^{14}\text{C}$, $^{13}\text{C}(\text{d}, \alpha_0)^{11}\text{B}$, $^{13}\text{C}(\text{d}, \alpha_1)^{11}\text{B}$ and $^{13}\text{C}(\text{d}, \text{t}_0)^{12}\text{C}$ nuclear reactions, that present different Q -values (respectively, 5.952, 5.169, 3.044 and 1.311 MeV), give rise to well separated peaks (respectively, at 5.9, 3.7, 2.3 and 1.3 MeV) with almost no background. The area under each peak, which corresponds to the number of emitted particles detected for a given solid angle, and the area of the gold peak can be directly used into Eq. (2) to calculate the corresponding cross section. The peak detected around 3 MeV corresponds to the $^{12}\text{C}(\text{d}, \text{p}_0)^{13}\text{C}$ nuclear reaction and allows us to characterize the carbon build-up phenomenon occurring during the analyses. This very slight carbon contamination increased linearly with the integrated charge and was determined to be

$(0.017 \pm 0.001) \times 10^{15}$ at $\text{cm}^{-2} \mu\text{C}^{-1}$. It represents a ^{13}C areal density increase lower than 10^{15} at cm^{-2} for a complete run of analysis (about 3000 μC) and was neglected for the determination of the differential cross section intensity.

The differential cross sections of the $^{13}\text{C}(\text{d}, \text{p}_0)^{14}\text{C}$, $^{13}\text{C}(\text{d}, \alpha_0)^{11}\text{B}$, $^{13}\text{C}(\text{d}, \alpha_1)^{11}\text{B}$ and $^{13}\text{C}(\text{d}, \text{t}_0)^{12}\text{C}$ nuclear reactions calculated from Eq. (2) for 135° , 150° and 165° laboratory detection angles are presented in Figs. 2–5 and will be available in the electronic IBANDL data base [9]. The absolute uncertainty on the cross sections is estimated at 4.5% in the energy range from 800 keV to 1650 keV and does not exceed 10% out of this range. It takes into account the uncertainties on the measurement of the ^{13}C foil composition (2.5%), the areal density of gold (2.0%), the number of emitted particles originating from the nuclear reactions under study (<3%) and the number of deuteron backscattered by gold nuclei (<0.5%). The energy scale was corrected in order to indicate the deuteron energy in

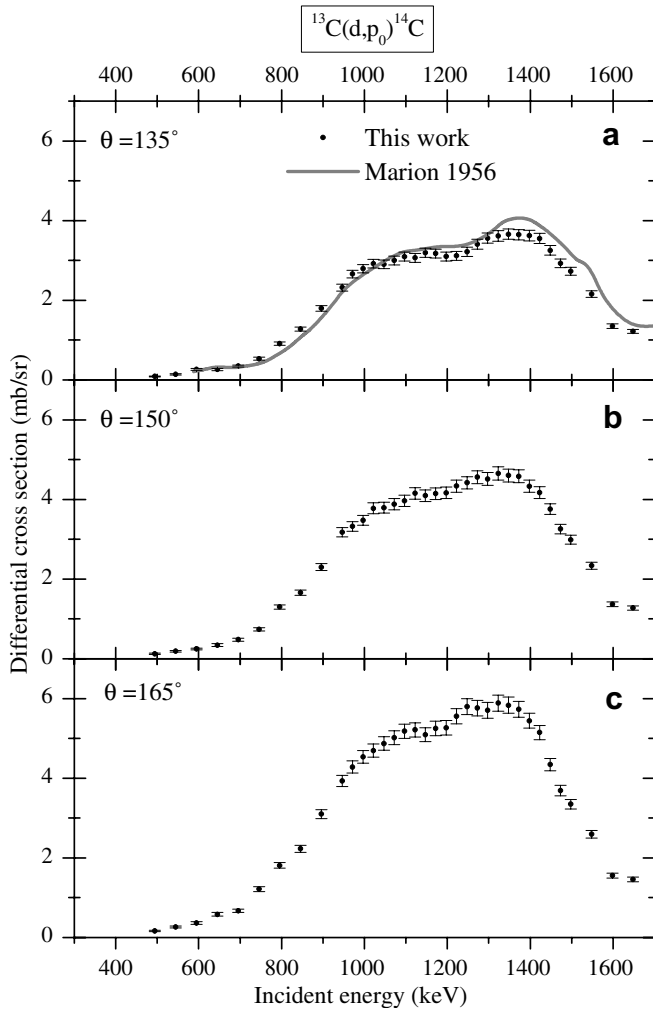


Fig. 2. Differential cross section curves for $^{13}\text{C}(\text{d}, \text{p}_0)^{14}\text{C}$ nuclear reaction measured at 135° (a), 150° (b) and 165° (c) in the laboratory system. Symbols (filled circles) represent the measurements performed in this work while the solid line represents measurements made by Marion and Weber [3].

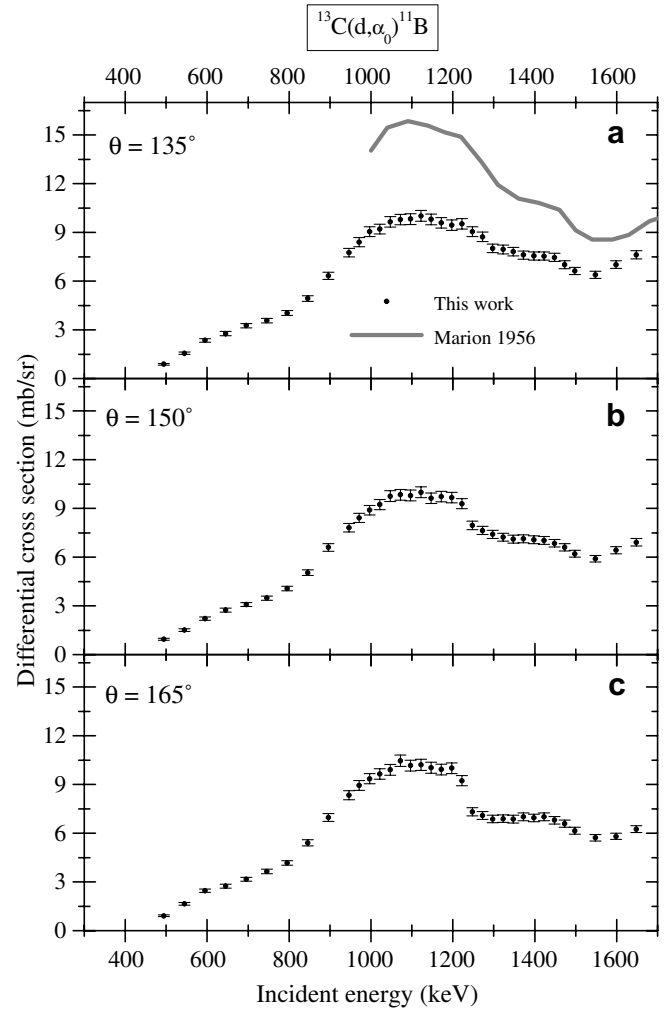


Fig. 3. Differential cross section curves for $^{13}\text{C}(\text{d}, \alpha_0)^{11}\text{B}$ nuclear reaction measured at 135° (a), 150° (b) and 165° (c) in the laboratory system. Symbols (filled circles) represent the measurements performed in this work while the solid line represents measurements made by Marion and Weber [4].

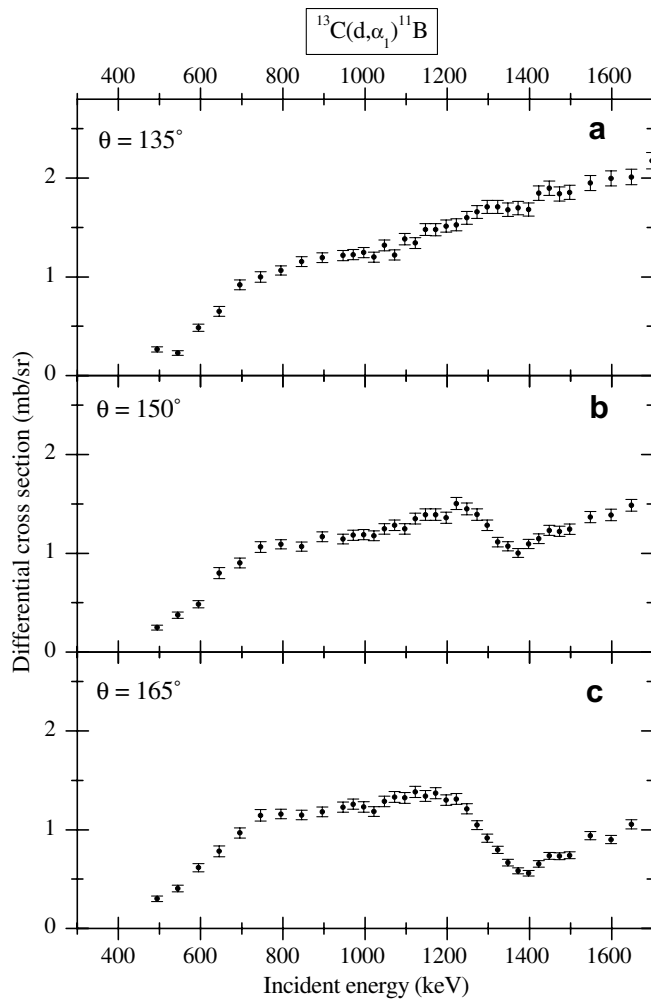


Fig. 4. Differential cross section curves for $^{13}\text{C}(\text{d}, \alpha_1)^{11}\text{B}$ nuclear reaction measured at 135° (a), 150° (b) and 165° (c) in the laboratory system. Symbols (filled circles) represent the measurements performed in this work.

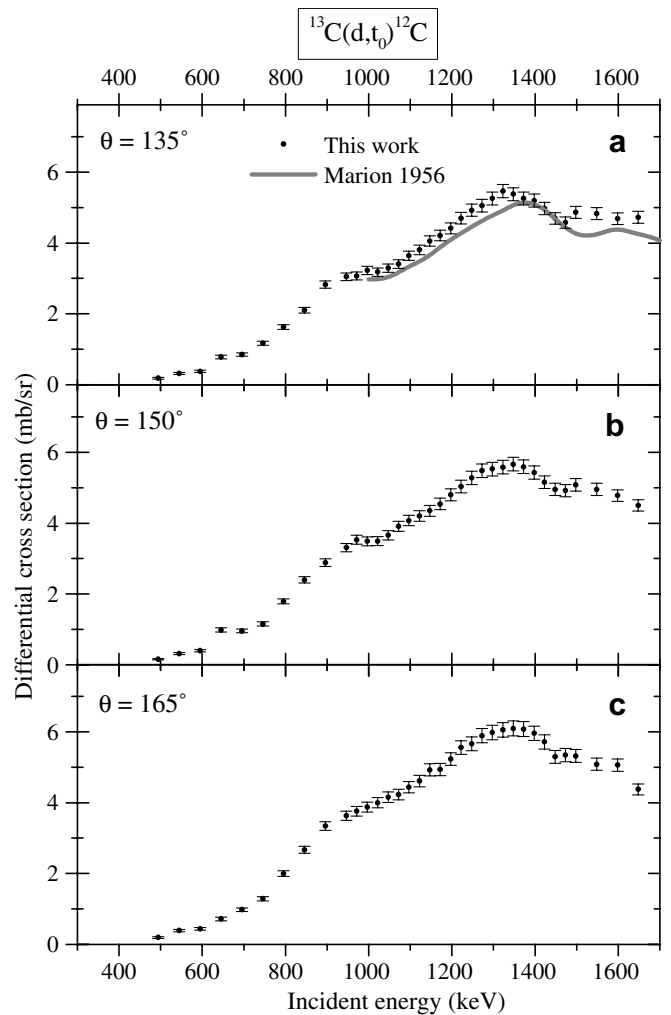


Fig. 5. Differential cross section curves for $^{13}\text{C}(\text{d}, t_0)^{12}\text{C}$ nuclear reaction measured at 135° (a), 150° (b) and 165° (c) in the laboratory system. Symbols (filled circles) represent the measurements performed in this work while solid line represents measurements made by Marion and Weber [4].

center of the thin carbon foil. This correction takes into account the energy lost by deuteron beam into carbon contamination layer, thin film of gold and thin ^{13}C foil. On the other hand, reference curves are uncorrected for target thickness used by Marion [3,4]. This is the reason why the cross sections measured in this work are systematically shifted of about 30 keV compared to those values (Figs. 2(a), 3(a) and 5(a)).

The $^{13}\text{C}(\text{d}, p_0)^{14}\text{C}$ cross section measured at 135° (Fig. 2(a)) is in very good agreement with the one measured by Marion and Weber [3]. The shape of the cross sections obtained at 135° for the $^{13}\text{C}(\text{d}, \alpha_0)^{11}\text{B}$ and $^{13}\text{C}(\text{d}, t_0)^{12}\text{C}$ reactions (Figs. 3(a) and 5(a)) is also in good agreement with the one from Ref. [4]. Nevertheless, we note significant differences in the intensity of these cross sections. This disagreement is due to the experimental procedure followed by Marion. Indeed, to facilitate the data acquisition with the magnetic spectrometer system [4], Marion has only measured the maximum of counting rate and not the area under the $^{13}\text{C}\alpha_0$ and $^{13}\text{C}t_0$ peaks. This method is correct at

high bombarding energy where the maximum of each peak is directly proportional to its surface. Nevertheless, at low deuteron energies the straggling of particles in carbon foil increases the width of the peak and its maximum is no more proportional to its area. In order to resolve this problem, a single empirical correction curve was established and applied to $^{13}\text{C}\alpha_0$ and $^{13}\text{C}t_0$ cross sections [4]. However, the nature and the energy of particles emitted in these reactions are very different. It is not correct to apply the same correction curve to the two cross sections. So the single empirical correction curve used by Marion leads to an overestimation of the $^{13}\text{C}(\text{d}, \alpha_0)^{11}\text{B}$ cross section of about 25% and to an undervaluation of the $^{13}\text{C}(\text{d}, t_0)^{12}\text{C}$ cross section of about 10%.

Finally, it is interesting to note that all nuclear reactions induced by deuteron on ^{13}C and on ^{12}C emit particles at well separated energies. Moreover these nuclear reactions present a cross section which does not vary much from 0.950 to 1.075 MeV. Thus it is possible to determine ^{13}C and ^{12}C depth distributions simultaneously with

a good accuracy working with a deuteron energy around 1.05 MeV.

4. Conclusion

A thin ^{13}C foil was used to measure differential cross sections of $^{13}\text{C}(\text{d}, \text{p}_0)^{14}\text{C}$, $^{13}\text{C}(\text{d}, \alpha_0)^{11}\text{B}$, $^{13}\text{C}(\text{d}, \alpha_1)^{11}\text{B}$ and $^{13}\text{C}(\text{d}, \text{t}_0)^{12}\text{C}$ nuclear reactions for deuteron energies from 0.5 to 1.65 MeV. These measurements were performed at 135° , 150° and 165° with respect to the incident beam. The ^{13}C areal density of the thin foil was measured by RBS using a 1 MeV ^4He beam. The build-up rate of carbon occurring during the analyses was evaluated and the contribution of ^{13}C enrichment to the thin foil was found to be negligible. The energy lost by the deuteron beam into this carbon contamination layer, the gold film and the thin ^{13}C foil was taken into account and the energy scale was corrected in order to indicate the deuteron energy in center of the thin carbon foil. Cross sections obtained for nuclear reactions at a laboratory detection angle of 135° are in quite good agreement with previous results in the literature [3,4]. The shapes of the cross sections are correct, but significant differences are observed in intensity for the $^{13}\text{C}(\text{d}, \alpha_0)^{11}\text{B}$ and $^{13}\text{C}(\text{d}, \text{t}_0)^{12}\text{C}$ cross sections. The usage

of nonresonant (d, p), (d, α) and (d, t) nuclear reactions allows to depth profile ^{12}C and ^{13}C in a single and relatively rapid measurement.

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